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THERMAL EXPANSION BEHAVIOR OF RANDOMLY ORIENTED SHORT CARBON FIBER REINFORCED COPPER COMPOSITES

Keiichi Kuniya, Hideo Arakawa and Takashi Namekawa

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16. Abstract

Several experiments were carried out on Cu composites with random orientation of short C fibers. The composite with random orientation of short C fibers swelled due to the elastic deformation of the fiber skeleton above 573 K. In a 3-dimensionally isotropic composite, the Cu matrix softened above 573 K and was fractured by the repulsive force of C fibers which were elastically deformed and released during heating. Swelling of the composite occurred when the fiber content was >20 vol. % or the fiber length exceeded 0.5 mm. The addition of carbide-forming elements such as Ti was effective in preventing the swelling.

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Thermal Expansion Behavior of Randomly Oriented Short Carbon Fiber Reinforced Copper Composites

Keiichi Kuniya*, Hideo Arakawa* and Takashi Namekawa*

We have already reported the development of a new copper-carbon fiber composite which possesses the properties of copper, i.e., the excellent electrical and thermal conductivities and the property of carbon fiber, i.e., a small thermal expansion coefficient[1]. The properties of the composite were expected to vary depending on the orientation of fibers. When applying the composite to some electronic devices, however, the properties of the composite have to be isotropic.

For this purpose, several experiments were carried out on the composites with random orientation of short carbon fibers. Results obtained in this study were as follows:

(1) The composite with random orientation of short carbon fibers swelled due to the elastic deformation of random-oriented carbon fiber's skelton at temperatures above 573 K. It was deduced that in a three-dimensionally isotropic composite such as above mentioned, the copper matrix softened above 573 K was fractured by the repulsive force of carbon fibers which had been elastically deformed and were released at heating, resulting in the swelling of the composite.

This swelling occurred, when the fiber content was over 20 volume percent or the fiber length exceeded 0.5 mm.

(2) It was found that the addition of carbide forming elements such as titanium was effective to prevent the above swelling.

(Received June 26, 1985)

Keywords: *short carbon fiber, copper carbon composite, random fiber composite, fiber reinforced metal, thermal expansion*

I. Introduction

The copper-carbon fiber composite uses copper on its matrix; thus it has high thermal and electric conductivity, and since it is reinforced by a carbon fiber which has a small and negative thermal expansion coefficient so that by adjusting its fiber contents the desired thermal expansion coefficient could be readily obtained. Accordingly, the copper-carbon fiber composites appear to be a suitable material for electric parts which necessitate high thermal and electric conductivity and have to have different thermal expansion coefficients by different combination of component materials [1-3].

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** Numbers in margin indicate pagination of foreign text.

Anisotropy develops in copper-carbon fiber composites in accordance with the direction of orientation of the fibers. However, isotropy is required when such composites are utilized in various types of electric parts. Various characteristics of 2-dimensional isotropic copper-carbon fiber composites utilizing long fibers have already been reported [1-7].

In general, 3-dimensional isotropy is obtained by random orientation of short fibers. However, from our experiments, it has been noted that such randomly oriented composites when subjected to high temperature tended to develop abnormal expansion due to exfoliation of the fiber-matrix interface. In this report trial modes of composites in which short fibers were randomly oriented in copper (hereinafter referred to as Cu-C short fiber composites) were constructed, and their thermal expansion behavior and relationship between the amount of carbon fibers and fiber lengths were studied. Also reinforcement of the fiber-matrix interface by the addition of Ti and Zr was studied.

II. Experimental method /1138

1. Raw material and method of constructing trial composites

For the fiber, the PAN type high strength carbon fiber in which 3000 unit fibers with a diameter of approximately 7 μm making up one bundle was used. The characteristics of this fiber were reported previously [1].

In addition to the copper used for the adjustment of the amount of carbon fibers, various elements with high carbide formation capability (with high negative values of changes in standard free energy on carbide formation) including Zr, Ti, Nb, V, W, Cr and Mo were selected in order to increase the interface strength of the fiber-matrix interface [8,9]. Average granular diameters of these elements in powder form were the greatest at about 62 μm for Cr and V, followed by 5-4 μm for Cu, Mo, T, Nb, then the smallest range of 2.2-1.6 μm for W and Zr.

Using these fiber bundles, about 1 μm of copper was placed on the surface of individual fibers by the method reported previously [1]. The fibers were cut into lengths of 0.5-10mm and stirred in water to form collective bodies of short fibers. Electrolytic copper powder and powders of added elements (Zr, Ti, Nb, V, W, Cr, and Mo) were adjusted to predetermined ratios, mixed and stirred in a 3% aqueous solution of methylcellulose, and Cu-X or X powder slurries were obtained. These Cu-X or X powder slurries, as will be noted below, were immersed and mixed with carbon fiber collective bodies. Due to the large difference in specific gravities of these powders and carbon fibers, the latter tend to be deflected. Then solvents with enough viscosity readily decompose and disperse after heating so that no residues remain in the composite were sought. It was noted that Cu-X or X powder slurries made with a 3% aqueous solution of methylcellulose when immersed in and mixed with the short fiber collective bodies, would result in the adhesion of copper and powder of added elemental powders on the surface of fibers, and even blending of fibers and added elements. Furthermore, it was also noted that methylcellulose is decomposed and disappears during hot pressing. Then this system was utilized.

Cu-X or X powder slurries adjusted to predetermined ratios are immersed with carbon fiber collective bodies in a vacuum mixed with a rotating mixer, and dried and solidifed at 375 K under atmospheric pressure. The solids thus obtained were hot pressed under N_2 + 8 vol % H_2 gas, with 24.5 MPa, 1273 K, and 1.2 k/s (20 min). The shape of the Cu-C short fiber composite obtained after hot pressing is 12 mm wide, 60 mm long and 4 mm thick. The apparent density of the testing composite differs according to the granular diameters of added elements. The composites to which Cr and V, with large granular diameters were added; these added elements deflected and the amounts of fibers were not uniform, and the apparent densities were about 80-95% of the theoretical values. In other cases, however, about 99% of the theoretical values were obtained.

2. Method of thermal expansion determination

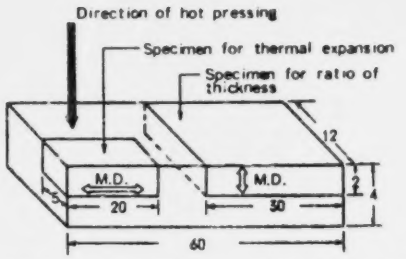

Figure 1. Schematic illustration of specimens (mm) and directions for characteristic measurements. (M.D. = measuring direction)

As shown in Figure 1, a piece of specimen 5 mm wide, 20 mm long, 2 mm thick, was taken and utilizing the differential type of thermal expansion measuring apparatus, the targeted thermal expansion rates and thermal expansion coefficients of Cu-C short fiber composites were computed by the difference between thermal expansion of the unknown testing specimen and that of the standard quartz sample. The direction of the measurement of the thermal expansion is right angle to the direction of the hot press, and the rate of fiber orientation is about 60%. Measurements under N_2 or He gas were made with the rate of heating at 0.067 k/s, the range of determination from room temperature to 1073 K. Along with the rate of thermal expansion, the mean thermal expansion coefficients from room temperature to 473 K was computed.

3. Method of determination of the rate of increase of plate thickness

In order to find the degree of abnormal expansion, samples of Cu-C short fiber composites, 12 mm wide, 30 mm long, 2 mm thick, as shown in Figure 1, were heated to 1073 K, 1.8 k/s (30 min) in hydrogen atmosphere, and the ratio of plate thicknesses before and after was obtained. This was designated as the rate of plate thickness increase. The direction of measurements of the rate of plate thickness increase was parallel to that of the hot press.

The condition of specimens and the layer of carbides of Ti and other elements was observed by an optical and scanning electron microscope, and was also linearly analyzed by an X-ray microanalyzer (EPMA).

III. Experimental results and discussion

1. Thermal expansion behavior of Cu-C short fiber composites

Figure 2 shows thermal expansion curves of Cu-C short fiber composites. As could be seen in the figure, specimen 1 with carbon fibers 30 vol %, shows the thermal expansion curve close to the hysteresis characteristic of fiber composites. In contrast, specimen 2 with carbon fiber 50 vol %, the thermal expansion curve shows linear expansion to around 473 K, followed by gradual expansion and a sudden increase is noted over 773 K. Observation after heating to 1073 K and cooling revealed the development of cracks in the specimen. Specimen 3, to which Ti was added, is shown for comparison (Cu-13.5 vol %, Ti-47 vol % C). As will be noted below, because this specimen had its fiber-copper matrix interface reinforced by the addition of Ti did not exhibit abnormal expansion and the hysteresis curve characteristic of fiber composites resulted.

Figure 3 shows the photomicrographs of specimen 2 before and after heating as seen by an optical microscope, and after heating as seen by a scanning electron microscope. By heat treatment at 1073 K, 1.8 k/s (30 min), carbon fibers and copper matrix were separated with formation of internal cracks and hollow cavities.

Figure 4 shows the rate of plate thickness increase for specimen 2 in Figure 2. The rate of increase accelerated at around 573 K with temperature elevation.

The following is offered as a possible factor in abnormal expansion seen in Cu-C short fiber composites. When fibers are cut into short segments they are oriented randomly. Fibers are entangled, forming a cotton-like skeleton. Since carbon fibers are elastic bodies, the skeletons formed are also elastic bodies. When these carbon fiber skeletons were pressed and enclosed within a copper matrix, a force of repulsion is internalized

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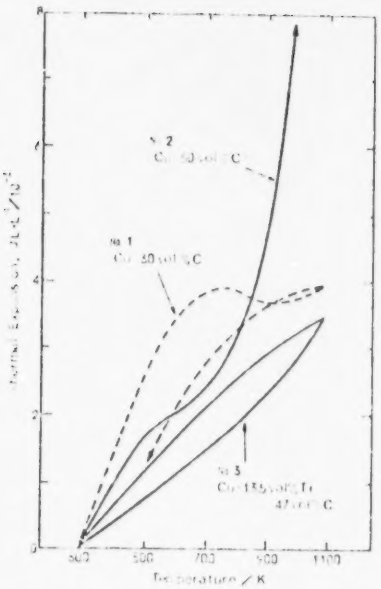

Figure 2. Effect of fiber volume contents and addition of Ti on thermal expansion of random composites containing 5 mm long carbon fibers.

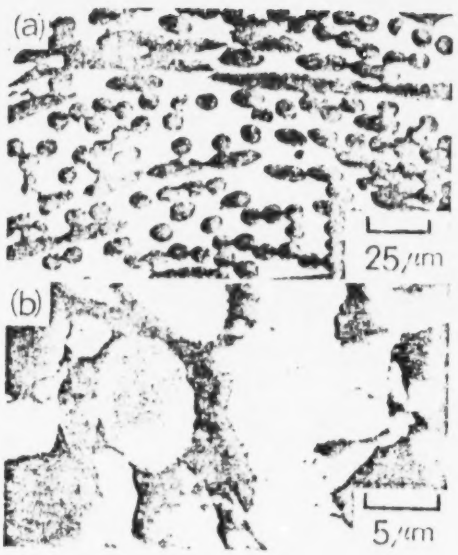

Figure 3. Microphotographs showing the effect of heat treatment on 5) vol% random fiber composites.
(a)--as hot pressed
(b)--after heat treatment at 1073 K for 1.8 ks.

with the composite. When heated under these conditions, carbon fibers maintain their strength and elasticity even under high temperatures so that with softening of the matrix, cracks are formed starting at the fiber-matrix interface, because of the force of repulsion of carbon fiber skeletons, eventually leading to destruction with the release of elastic energy.

On the other hand, thermal expansion curves of composites frequently made with a combination of materials with different characteristics exhibit hysteresis curves as seen with the Cu-W fiber composite [10]. The area surrounded by this curve depends on the difference of characteristics of component materials of the composite. With greater area, greater thermal stress is generated [11,12]. With Cu-C short fiber composites, the large difference between the thermal expansion coefficients of copper

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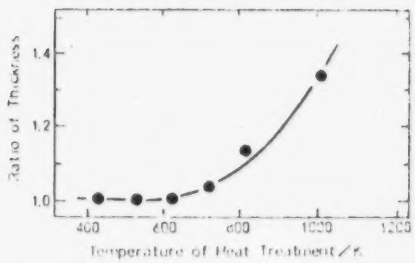

Figure 4. Effect of temperature on ratio of thickness after heat treatment to that before heat treatment of 50 vol% random fiber composite.

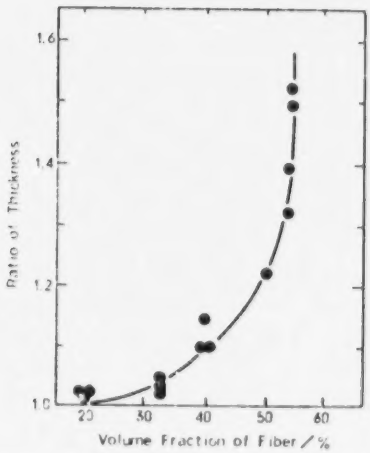

Figure 5. Ratio of thickness after heat treatment to that before heat treatment at 1073 K for 1.8 ks, vs. volume fraction of 5 mm long fiber in random composites.

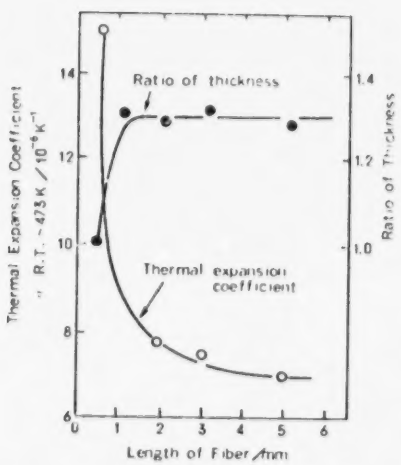

Figure 6. Thermal expansion coefficient (R.T.-473K) and ratio thickness after heat treatment to that before heat treatment at 1073K for 1.8 ks, vs. length of fiber in 50 vol% random composites.

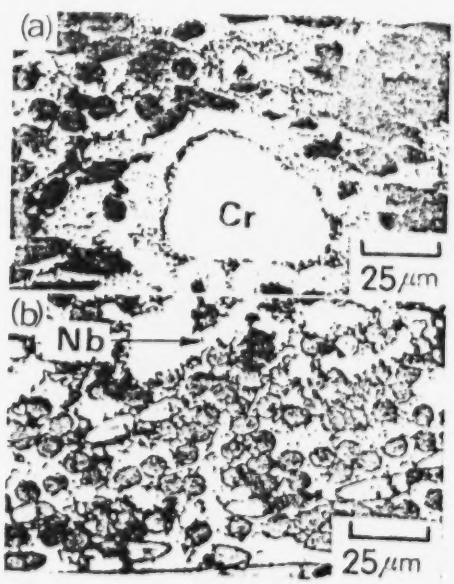

Figure 7. Microphotographs of random composites, as fabricated, containing several carbide forming elements.
(a) Cu-17.3 vol%Cr-44.0 vol%C
(b) Cu-6.7 vol%Nb-49.0 vol%C

and carbon fibers suggests that a very large amount of thermal stress is generated. However, there is no moisture between the fibers and copper matrix, and the copper is merely adherent physically or mechanically as the surface of fibers. Accordingly, the force of bonding between the fiber and matrix is small, and as thermal stress increases, the fiber-matrix interface becomes more easily separated.

Thus, abnormal expansion is thought to be caused mainly by the elastic energy of fibers becoming greater than the adhesion strength of the fiber-matrix interface, to which thermal stress generated by the difference in thermal expansion between copper matrix and carbon fibers is added, leading to the separation of the fiber-matrix interface, and formation of hollow cavities.

2. Effects of the amount and length of fibers on thermal expansion characteristics

The relationship between the rate of plate thickness increase and carbon fiber contents is shown in Figure 5. The length of the fiber used is 5 mm. As can be seen from the figure, abnormal expansion does not occur when the amount of carbon fiber is 20 vol% or less.

Figure 6 shows the relation between the length of carbon fibers and the rate of increase of plate thickness. When the fiber length is less than 0.5 mm, abnormal expansion does not take place but thermal expansion coefficient increases.

In order to have a functional fiber reinforced mechanism in short fiber composites, it is necessary for the fiber to have the aspect ratio greater than the critical level. This varies with different fibers, but is generally around 100. On the other hand, in Cu-C short fiber composites, the thermal expansion coefficient decreases because the fiber reinforced mechanism works, and the strength of the copper matrix is inhibited by fibers.

When the fiber length is 0.5 mm, the fiber diameter is 7 μm, so that the aspect ratio is about 70, and with the fiber length below 0.5 mm, it becomes less than the critical aspect ratio. Accordingly, while abnormal expansion does not take place because the effects of fiber reinforcement mechanism becomes smaller, but the thermal expansion coefficients also do not become smaller.

3. Effects of carbide forming elements on thermal expansion characteristics

Figure 7 shows optical photomicrographs of Cu-C short fiber composites to which Cr, with a large granular diameter, and Nb, with a small granular diameter, are added. The added elements are dispersed in the state of the original powder. The reason that these elements are present in the state close to that of the original powder is that most of these elements do not melt into the copper matrix and do not form compounds [13]. Accordingly, the only elements in contact with carbon fibers take part in reaction, and those elements present in distance from carbon fibers do not get involved in reaction. For example, a semicircular layer of the reaction is noted in carbon fibers around Cr.

Figure 9 shows the type of elements, the relationship of the amount added and the rate of plate thickness increase in Cu-C short fiber composites. As could be seen from the figure, greater effects are noted with Ti, Zr and Nb. Especially when more than 5 vol% of Ti and Zr are added, a marked decrease in the rate of plate thickness is noted.

The following general tendency is said to exist in terms of melting properties of various melted metals to carbon or carbide [14]. When various elements are added to a melted metal, the melting contact angle between the metal and the carbon varies with different elements added, but elements with a high capability of carbide formation (those elements with a high negative value of free energy of a carbide formation) cause a greater decrease

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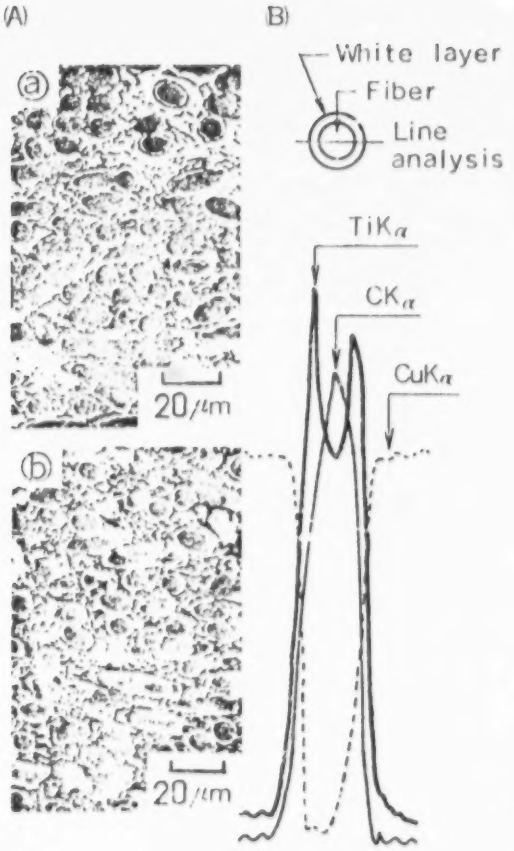

Figure 8. Microphotographs of random composites containing Zr or Ti, and line analysis data by EPMA for the specimen (b).
(A) Microphotographs
(a) Cu-8.7vol%Zr-47.0 vol%C.
(b) Cu-13.5vol%Ti-47.0 vol%C.
(B) Line analysis of EPMA

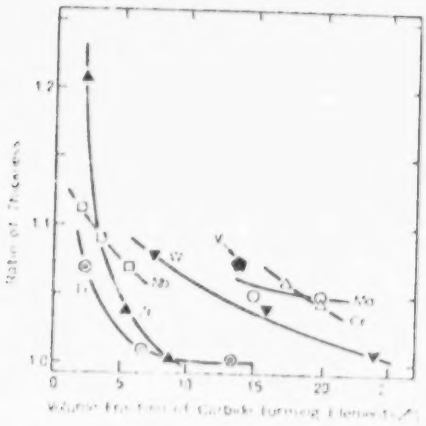

Figure 9. Ratio of thickness after heat treatment to that before heat treatment at 1073 K for 1.8 ks, of random composites as a function of the amount of additives.

of the angle of contact with a less amount, improving the melting property.

When Ti and Zr are added, at certain composition ratios, the matrix melting points become less than the hot press temperature (1273 K±50K) due to a co-crystallization reaction [13]. Accordingly, portions of the matrix become melted, and since these elements have a high carbide formation capability, melting properties between the matrix and the fiber are improved, forming a layer of carbides on the fiber surface. Simultaneously, these are dispersed over the interface between the matrix and the carbide, reinforcing the bonding between the matrix and the carbide. Such an improvement in the bonding strength of the carbon fiber-matrix interface is thought to lead to the inhibition of an increase in the plate thickness from heating.

IV. Conclusion

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Thermal expansion behavior of Cu-C short fiber composites in which carbon fibers cut to short lengths were randomly oriented in copper, was studied and the following findings were obtained.

(1) When exposed to a temperature above 573 K, Cu-C short fiber composites developed a separation of the fibers and copper matrix, forming hollow cavities which resulted in bulging. This abnormal expansion was mainly due to the elastic stress of a short fiber skeleton that exists within the matrix and was seen when the amount of fiber was over 20 vol%, or the length was over 0.5 mm.

(2) The abnormal expansion noted above could be stopped by adding elements that form carbides, such as Ti, Zr, Nb, to the copper matrix. This was thought to be due to the formation of carbides of elements such as Ti, increasing the strength of the carbon fiber-copper matrix interface.

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